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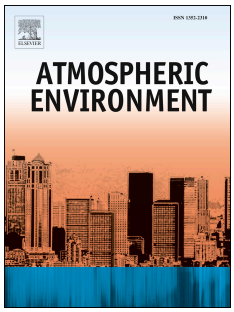

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EMISSION OF ULTRAFINE PARTICLES FROM THE INCINERATION OF MUNICIPAL SOLID WASTE: A REVIEW

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HIGHLIGHTS

- Incineration is increasing as a waste disposal option
- Atmospheric emissions are an important concern
- Abatement plant is highly efficient for particulate matter
- Ultrafine particle emissions are generally very low

ABSTRACT

Ultrafine particles (diameter <100 nm) are of great topical interest because of concerns over possible enhanced toxicity relative to larger particles of the same composition. While combustion processes, and especially road traffic exhaust are a known major source of ultrafine particle emissions, relatively little is known of the magnitude of emissions from non-traffic sources. One such source is the incineration of municipal waste, and this article reviews studies carried out on the emissions from modern municipal waste incinerators. The effects of engineering controls upon particle emissions are considered, as well as the very limited information on the effects of changing waste composition. The results of measurements of incinerator flue gas, and of atmospheric sampling at ground level in the vicinity of incinerators, show that typical ultrafine particle concentrations in flue gas are broadly similar to those in urban air and that consequently, after the dispersion process dilutes incinerator exhaust with ambient air, ultrafine particle concentrations are typically indistinguishable from those that would occur in the absence of the incinerator. In some cases the ultrafine particle concentration in the flue gas may be below that in the local ambient air. This appears to be a consequence of the removal of semi-volatile vapours in the secondary combustion zone and abatement plant, and the high efficiency of fabric filters for ultrafine particle collection.

Keywords: Ultrafine particles; nanoparticles; particle removal devices; incinerator emissions

1. INTRODUCTION

The regulation of municipal solid waste (MSW) incineration plants within the European Union was strengthened by the Waste Incineration Directive, 2000/76/EC (WID), which applied to new plants from December 2002, and to existing plants from December 2005 (EU, 2000), resulting in the closure or upgrading of many of the older installations. Further WID requirements on the incineration of MSW have been carried into the Industrial Emissions Directive 2010/75/EU (EU, 2010). The daily average air emission limit for total dust within the exhaust gases is set at 10 mg m^{-3} (EU, 2010), while emission limits are also set for certain metals and their compounds (including their gaseous forms). The specified metals are cadmium and thallium (the sum of which is limited to 0.05 mg m^{-3}), mercury (limited to 0.05 mg m^{-3}), and antimony, arsenic, lead, chromium, cobalt, copper, manganese, nickel and vanadium (the sum of which is limited to 0.5 mg m^{-3}). Limits are also set on various gaseous pollutants, including gaseous and vaporous organic substances (expressed as total organic carbon, TOC), and dioxin and furan emissions.

Although ambient air quality standards for particulate matter are set in terms of the permissible mass concentrations of certain ranges of aerodynamic size of particle (EU, 2008), there has been debate in recent years (e.g. Seaton et al., 1995; Lighty et al., 2000; Delfino et al., 2005) as to whether smaller particles have a more deleterious effect on human health, and ambient air quality standards would be better if written in terms of the particle number concentration, reflecting the concentration of "ultrafine", defined as those particles with an aerodynamic diameter of less than 0.1 µm (100 nm) (Lighty et al., 2000). Concerns have been raised (e.g. Allsopp et al., 2001; AIE, 2008) as to whether the regulation of MSW incineration plants by a mass based standard overlooks the effects of the smaller particles that may be emitted from these plants, and that metals may be present at enhanced concentrations in these smaller particles (Zhang et al., 2008). The recent development of engineered nanomaterials has also raised the question of the consequences of the disposal of products containing these materials by incineration (Holder et al., 2013).

A limited literature exists on the particle number concentrations and size spectra of emissions from MSW incineration plants in contrast to studies of particle mass concentrations (e.g. Ragazzi and Rada, 2012). Reviews of the literature have been carried out by Yinon et al. (2010) and Yinon (2010) who found that MSW incineration made a minor contribution to ultrafine particles in the United States compared to coal combustion, and by Le Cloirec (2012) who described the operation of a number of MSW incineration plants and the range of flue gas cleaning techniques used in Europe. Holder et al. (2013) reviewed the issue of the incineration of engineered nanomaterials finding a limited number of theoretical studies, but only a single study in which a full size MSW incineration plant was used experimentally (Walser et al., 2012).

The purpose of this review is to identify reports of measurements of ultrafine particulate in the flue gases from MSW incinerators, and to assess their relevance to the operation of MSW incinerators in the United Kingdom.

2. METHODOLOGY

For the purpose of this review, the "Web of Science" and "Google Scholar" databases were searched using the terms;

"waste" and "incineration" and "emission" and ("ultrafine" or "$PM_{0.1}$" or "PM_1"),

over the period 2000 to January 2016 to find papers reporting measurements of ultrafine particulate matter in the flue gases from MSW incinerators, and further references were identified from the papers thus located. Few papers were found via either "Web of Science" or when "Google Scholar" was limited to words in the title. Several thousand were found by "Google Scholar" when words in the text were included, largely as a result of many authors identifying waste incinerator emissions as a possible source of particulate matter. The majority of papers cited below were identified by

forward and backward citations. The starting date of 2000 was chosen because of the major changes in the design and operation of MSW incinerators in Europe following the implementation of Directive 2000/76/EC.

3. BACKGROUND

3.1 Number vs. Mass Measurement of Particulate Matter

Airborne particulate matter consists of particles covering a wide size range which can be quantified in terms of either the mass (or volume) or number of particles within a unit volume of air (Harrison et al., 2000). A consequence of the wide range of particle sizes is that measures of particle number concentration are dominated by the smaller sized particles, while as a result of particle mass varying with the cube of particle diameter, measures of particle mass concentration are dominated by the larger sized particles, as shown generically in Figure 1, which presents the particle number and particle size distributions in rural air for typical ambient conditions. Traditionally, mass concentration measurements have been used to assess air quality, although often with an upper limit on the size of particle considered as larger particles are unlikely to reach the human lung and therefore have limited health effects. For example, PM_{10} is the mass concentration of particles with an aerodynamic diameter of 10 µm or less, where the aerodynamic diameter is the diameter of a spherical particle with density of 1 g cm^{-3} with the same terminal settling properties.

Particles are lost from the atmosphere by sedimentation and impaction in the case of larger (> 1 µm) particles, and by coagulation due to diffusion and electrical effects in the case of smaller (< 0.1 µm) particles. Intermediate sized particles tend to have a longer lifetime in the atmosphere, resulting in an increase in the concentration seen in both number and mass spectra between 0.1 and 1.0 µm (Figure 1) , known as the “accumulation mode”.

These attributes also result in this particle size range (0.1 – 1 µm) being the most difficult to capture in filtration systems (Zeuthen et al., 2007). Larger particles may be captured by interception (where a particle following the flow is within the particle radius of a filter element), impaction (where the inertia of a particle carries it onto a filter element), and - to a limited extent – gravitation (where a particle settles on a horizontal element of the filter) (Hinds, 1999). Smaller particles may be captured by diffusion (where the particles are moving relative to the flow) and both larger and smaller particles may be captured by electrostatic effects if either the particle or the filter elements are charged. The combination of these processes tends to result in minimum filter efficiencies at particle diameters around 0.2 µm, although when the filter elements carry a charge the minimum efficiency is improved, and tends to occur at a lower particle diameter (Brown, 1993).

Larger particles generally enter the atmosphere through mechanical abrasion processes or through resuspension. Smaller particles are frequently formed through nucleation - the condensation of semi-volatile substances, often originating from combustion processes, when the vapour concentration reaches saturation. However, in general, condensation preferentially occurs on the surfaces of existing particles (Lighty et al., 2000), rather than causing the formation of new particles. In the atmosphere, the occurrence of high concentrations of nucleated particles may be evidence of a low concentration of larger particles (Charron et al., 2007; Cusack et al., 2013). Smaller particles are also formed as soot in fuel rich conditions by the agglomeration of, and chemical reactions between, hydrocarbon fragments (Lighty et al., 2000).

The dependence of particle formation mechanisms on the temperature and degree of saturation of vapours may result in the concentration of particles measured being influenced by conditions in the exhaust flue. The sampling of particulate matter from hot flue gas requires the gas to be diluted and cooled under controlled conditions. While the combined mass concentration of gases and particles is conserved, this is not the case for particle number concentration, and differences in dilution

technique may make it difficult to compare the particle size concentrations measured by different investigators (Lightly et al., 2000). This issue is also encountered in the measurement of particulate emissions from vehicle engines where exhaust conditioning and dilution processes are widely used (Burtscher, 2005; Shi and Harrison, 1999).

3.2 MSW Incineration Operations

The products of combustion from MSW incineration plants will depend upon the design of the plant, conditions within the combustion chamber, and the components of the materials being burned, which in the case of municipal solid waste can be diverse, and may vary over short periods of time. In the presence of high concentrations of chloride, metals will form chlorides (Jiao et al., 2011; Jiao et al., 2013) rather than oxides, while metals may themselves act as catalysts in other reactions (Pena et al., 2012). Within the lower temperature exhaust gases dioxins and furans may be formed (Vehlow, 2012) and combustion and after-treatment conditions are normally adjusted to minimise their formation.

Contact of the combustion gases with specific chemicals added in the treatment system is used to reduce emissions of prescribed components. Lime, hydrated lime, or sodium bicarbonate can be used to neutralise acid gases (Le Cloirec, 2012), and in the case of a sodium based sorbent can also partially reduce NO_x concentrations (Verdone and De Filippis, 2004). The introduction of activated carbon will result in the adsorption of dioxins and furans and volatile metals such as mercury (Le Cloirec, 2012). NO_x emissions can be reduced by the introduction of ammonia or urea within a system applying either selective catalytic reduction (Le Cloirec, 2012) or selective non-catalytic reduction (SNCR) as used in the United Kingdom. The various techniques for the control of air pollution from MSW incinerators, and the history of their development, are reviewed by Vehlow (2015).

The size of particulate matter within the incinerator tends to be relatively large compared to oil or gas fueled combustion systems. Maguhn et al. (2003) measured a maximum concentration within the particle size spectrum before flue gas treatment at around 100 nm diameter when back-up oil burners were switched off, and an additional second mode at around 30 nm diameter when the burners were switched on. The introduction of absorbent and adsorbent materials into the process will increase the particulate mass in the exhaust gases, and increase the need for particle removal systems. Commonly used methods of removing particles from the waste gas stream are cyclones, electrostatic precipitators and, on most recent MSW incineration installations, fabric (or bag-house) filters.

MSW incineration plants with energy recovery which are currently operational within the United Kingdom are reviewed by Nixon et al. (2013) who note that the flue gas treatment processes and technologies on current UK incinerators are all relatively similar. Some plants use recirculation of the flue gases in the combustion chamber to reduce emissions of NO_x and injection of urea and/or ammonia to the combustion chamber for further reduction of NO_x emissions. Either hydrated lime or lime milk is injected into the flue gases exiting the combustion chamber to remove sulphur dioxide and hydrogen chloride in a dry or semi-dry gas scrubbing system. Activated carbon is also injected into the flue gases to adsorb vapour phase dioxins, furans and volatile metals. The gas is then passed to a fabric filter system and from thence to the chimney stack. A schematic diagram of the process is shown in Figure 2.

Particles arise in the incinerator processes from the entrainment of ash particles into the flue gases exiting the furnace as well as from the condensation of vapour phase materials as the flue gases cool. The vast majority of particles will have formed by the time the particles enter the cooler environment of the emissions abatement system. As outlined below, it is possible for further

particles to form subsequent to the passage of exhaust gases through the emissions abatement system, but in normal configurations of plant, this is likely to be rather insignificant.

3.3 Factors Influencing the Efficiency of Particle Removal by Fabric Filters

In modern MSW incinerators in the United Kingdom the final gas cleaning stage in a waste incinerator is a fabric filter (bag filter) which collects not only particles produced in the incinerator but also the chemicals added in the emissions abatement process. These filters are far more efficient than would be anticipated from a simple sieving mechanism (referred to as interception), and are far more efficient than other particle collection systems such as cyclones and electrostatic precipitators (Lighty et al., 2000). This is because within a fabric filter particles are collected by five separate mechanisms; interception, inertial impaction, diffusion, gravitational settling and electrostatic attraction (Hinds, 1999). The processes of interception, impaction and gravitational settling are efficient for coarse particles while ultrafine particles are efficiently removed by the mechanisms of diffusion and electrostatic attraction. For this reason, filters are typically very efficient for particles greater than 0.8 µm diameter and for those smaller than 0.1 µm diameter as measured by Yi et al. (2008). Collection efficiencies can be improved, and the diameter of minimum efficiency reduced, where there is an electrical charge on the filter elements (Brown, 1993). The build-up of particles on the filter surface during operation has the effect of adding to the depth of the filter and consequently enhancing its efficiency. Because of this combination of mechanisms, the fabric filters used in municipal waste incinerator plants are expected to be highly efficient for the removal of ultrafine particles. The high collection efficiencies and low diameter of minimum efficiency may be evidence of a build up of electrical charge on the filter elements and collected material.

4. REVIEW OF PUBLISHED DATA

4.1 Measured Emissions – Particle Number

Measurements of particle number concentrations, or of sub-micrometre particle number size distributions, of emissions from MSW incinerators fitted with fabric filters have been reported by a limited number of authors. Studies involving 17 MSW incineration plants are listed in Table 1, along with plant operational details. Where the result for more than one plant is reported in any paper, these are numbered separately in a manner similar to that in the original paper. Although in several cases the authors report concentrations measured at various stages through the incineration process, the value of particle number concentration given in Table 1 is that for the final (and lowest temperature) measurement stage.

In reviewing measurements of ultrafine particles in emissions from a combustion source, it is important to recognise that the sampling protocol can influence the measured number concentration and size distribution. An extreme case is that of diesel engine exhaust where the use of low dilutions has a tendency to suppress the formation of newly nucleated particles derived from semi-volatile constituents of the exhaust gases (e.g. Shi and Harrison, 1999). Generally speaking, the use of high dilution with cool clean air prior to particle measurements will tend to increase the number concentration of particles when calculated back to a concentration in undiluted exhaust gas, although the mass concentration of particles is not significantly affected, the mass of particles and vapour being conserved. In the case of diesel exhaust, new ultrafine particles can be formed from condensation of semi-volatile vapour of lubricating oil present in the exhaust gases.

New particle formation is dependent upon the concentrations of condensable vapours and pre-existing particle surface area and will therefore be influenced by the dilution and temperature of the gases when measurements are made. The measurement temperature and dilution conditions (where available) are given in Table 1. The results from Cernuschi et al. (2012) are the highest particle

number concentrations measured after the fabric filter at each plant, which are those obtained with highest dilution (except in the case of plant 4 (I) where higher concentrations were recorded with medium dilution), and where the diluted stream temperature was between 24 – 31^{0}C. Examples of the particulate size distributions measured in the emissions of incinerators by a number of authors are shown in Figure 3.

Buonanno et al. (2012) measured particulate matter from the flue gas after a thermodilution unit within a system designed to control condensation and nucleation processes, and corrected the results for diffusion losses. Wilen et al. (2007) reduced the sample flow temperature to 20 – 50 ^{0}C in a porous tube diluter, but does not give a figure for the dilution ratio. Maguhn et al. (2003) and Zeuthen et al. (2007) give ranges over which the sample could be diluted but are not specific as to what dilution was used. Dilution ratios are generally calculated by measuring carbon dioxide concentrations in the flue and in the sample measured. FORCE (2009) and Fuglsang et al. (2010) report particle number concentrations measured at three MSW incineration plants, including one which had an electrostatic precipitator and agglomeration filter rather than a fabric filter which is why it is not considered in Table 1. The particle number concentration measured by FORCE (2009) at plant WTE3 (Table 1, N) is exceptionally low. The authors suggest that this may be due to the combined use of an electrostatic precipitator in addition to a fabric filter, and also note that the mass size distribution was dominated by particles with diameter greater than 2.5 µm. Even if the exceptionally low measurement by FORCE (2009) (Table 1, N) is disregarded, the measured particle number concentrations of the emissions in Table 1 are generally low compared to typical ambient particle concentrations measured in London (Table 2) (NPL, 2013). Ozgen et al. (2012) suggest that the higher particle number concentrations measured on the plant with a wet particulate collection system may be due to the operation of that system.

It should be noted that many of the experimental studies on particle number concentrations of emissions from MSW incinerators were conducted on Italian plants that had low total particulate matter emissions (Table 1). In a study of one Danish MSW incinerator and one Italian MSW incinerator, Turconi et al. (2013) found that while the Danish system had the better overall environmental performance, flue gas cleaning was better at the Italian plant. While all measurements of ultrafine particulate matter considered in this report were obtained from plants operating within the EU regulatory framework, there may be differences in operational practices and in the waste processed that would affect emissions.

4.2 Effects of Combustion Processes on Particle Emissions

The number and size distributions of the particles emitted by incineration will depend upon the materials burnt and the combustion processes within the incinerator, and upon any device inserted into the exhaust gas stream to remove particulate matter.

In addition to measurements made on typical operational waste, Zeuthen et al. (2007) and Pedersen et al. (2009) introduced additional quantities of selected waste materials and measured the particulate matter before flue gas treatment to examine the effect of different waste types on ultrafine particulate production. The additional wastes were; PVC plastics (high organically bound chloride), vehicle disposal waste after recyclables removal (high chloride, alkali metals and heavy metals), batteries – excluding car batteries - (high heavy metals), treated wood waste (high copper, chromium, arsenic), shoes (high chromium) and de-icing salt (high inorganic chloride). The largest increases in both mass ($PM_{2.5}$) and number concentrations (of around 50%) were obtained with the additional de-icing salt. Iron and copper were found to be major elements in the smaller particles when vehicle disposal and battery waste were added but in the case of larger particles (> 100 nm) were less than 1% of the total weight, a measurement that was not considered reliable by Zeuthen et

al. (2007). Pedersen et al. (2009) concluded that the concentration of inorganically bound chloride in the waste was positively correlated with the concentration of alkali metals in the fly ash.

A comparison of grate-firing and circulating fluidised bed waste combustion plants (Lind et al., 2007) found that at measuring points before fabric filter inlets the latter had a lower concentration of sub-micron particulate matter, 0.30 g m^{-3}, compared to 1.1 g m^{-3} for the grate-firing plant. There was also a difference in modal size of the particles, with average modal diameters of 0.4 µm and 0.6 µm respectively for the fluidised bed and grate-fired plants. Problems with sampling prevented reliable results being obtained for larger (>2.5 µm) particles at the fluidised bed plant. Lind et al. (2007) suggest that the difference between the two plants is that due to higher peak temperatures in the grate-fired plant, a larger proportion of ash-forming compounds was released to the gas phase during combustion before nucleation and condensation to form fine particles. In the fluidised bed plant, the authors propose that the lower temperature resulted in the release of less ash-forming compounds, with those compounds that were formed then reacting with coarse fly ash and the larger particles of the fluid bed. The use of grate firing systems for biomass fuels is reviewed by Yin et al. (2008).

The propensity for heavy metals to volatilise during combustion depends upon the presence of other elements (Yao and Naruse, 2009). The presence of chloride reduces the temperature at which condensation of heavy metals such as Pb, Cd, Cr and Hg occurs, hence increasing their volatility, while the presence of sulphur increases the temperature of condensation and reduces the volatility of the metals (Yao and Naruse, 2009).

The effect of the introduction of silicon-aluminium based sorbents into the combustion process has been examined by Linak et al. (2003) and Yao and Naruse (2009). The introduction of kaolin resulted in reductions in the concentrations of vanadium, nickel, iron and zinc (in particles < 0.56

329 µm) of respectively 35%, 56%, 56% and 40% (Linak et al., 2003), while lead and cadmium
330 concentrations were reduced by 40% - 45% (Yao and Naruse, 2009). In the number concentration
331 distribution, there is a reduction in the number of particles at diameters less than 0.1 µm with an
332 increase at larger diameters (Yao and Naruse, 2009), while particle mass is reduced at particle
333 diameters around 0.08 µm to 0.5 µm, and increased at diameters around 1 µm to 10 µm (Linak et
334 al., 2003; Yao and Naruse, 2009). Yao and Naruse (2009) found that zeolite was slightly less
335 effective than kaolin, and that in general silicon-aluminium sorbents performed better than calcium
336 based sorbents in capturing lead and cadmium. The processes of metal vapour adsorption by
337 kaolinite is discussed by Gale and Wendt (2003) and Yoo et al. (2005).
338
339 In a study on the heavy metal concentrations in the furnace ash and the fly ash collected by the fabric
340 filters of two MSW incinerators in Shanghai, Zhang et al. (2008) were able to distinguish between
341 those metals (Cr, Cu and Ni) which were not easily volatilised, and were transferred to the fine
342 particulate matter that was collected in the fabric filters by entrainment and therefore had smaller
343 mass concentrations in sub 30 µm particles than in 74 – 150 µm particles in the case of chromium
344 and nickel while copper concentrations were twice as high in sub 30 µm particles than in the 74 -150
345 µm size range. In contrast, those metals (Hg and Cd) which were easily volatilised and were
346 transferred to the fine particulate by an evaporation-condensation-adsorption process and therefore
347 concentrations in the sub 30 µm particles which were four times those occurring in 74 – 154 µm
348 particles. Lead and zinc which were transferred to the fine particulate by both entrainment and
349 evaporation had a more gradual increase in concentration with reducing particle size. Although all
350 heavy metals had a higher concentration in the particulate matter collected by the fabric filters than in
351 the furnace ash, the larger quantity of furnace ash resulted in the majority of all metals being found in
352 the latter. The ratio of metal in the fly ash to furnace ash was greater for the more volatile metals.
353 These incinerators were operated with lime slury ($Ca(OH)_2$) to remove acid gases, activated carbon to

remove metals and dioxins, and final fabric filters to control particulate matter emissions. The plants operated at a temperature of about 850^0C, and burnt approximately 1200 and 1500 t day^{-1}.

Unlike diesel engines where significant numbers of ultrafine particles can be formed by condensation in the exhaust, due to the introduction of activated carbon in a MSW incinerator to remove semi-volatile constituents (such as some dioxins and furans) combined with a high burn-out from the combustion process, the after-treatment will tend to result in extremely low concentrations of semi-volatile constituents and consequently there will be little potential for new particle formation during incinerator combustion gas dilution. There may be an exception (mentioned later) where aqueous scrubbing is conducted subsequent to other after-treatment processes which may promote new particle formation from inorganic precursors such as sulphur dioxide. Such situations are, however, unusual.

4.3 Combustion of Engineered Nanomaterials

Holder et al. (2013) identified a limited number of papers which considered the effect of municipal waste incineration on the release of engineered nanoparticles from materials containing them, and only one experimental study (Walser et al. 2012) on a full sized MSW incinerator.

Roes et al. (2012) conducted a preliminary assessment of the fate of engineered nanoparticles during MSW incineration and suggest that removal systems are inadequate to deal with bulk quantities of nanomaterials in municipal waste. This assessment however, assumes that no engineered nanoparticles are destroyed by incineration, and appears to be based on an illustrative figure of the capture efficiency of respirators contained in Centres for Disease Control and Prevention guidance on N95 respirators and surgical masks (CDC, 2009), which is not a quantitative report of the efficiency of either N95 respirators and surgical masks or of the fabric filters used on modern MSW incinerators. The filtration media used in personal respirators are

frequently charged and collection efficiencies may differ from those of the fabric filters used on MSW incinerators. A theoretical study of the fate of engineered titanium oxide, zinc oxide, silver nano particles and carbon nanotubes by Mueller et al. (2013) assuming (rather optimistic) particle removal efficiencies of 99.995% by an electrostatic precipitator concluded that 94% of carbon nanotubes would be destroyed by incineration, with other nanomaterials ending up within the incinerator bottom ash or being captured by the flue gas control systems, with less than 0.0001% being emitted to the air.

Experimental trials were conducted by Walser et al. (2012) in which cerium oxide nanoparticles (80 nm modal diameter) which are expected to be stable in incineration, were introduced into a MSW incinerator (10 kg in the waste feed; or 1 kg into the furnace) fitted with an electrostatic precipitator and a wet scrubber, and the quantity of cerium oxide in the bottom ash, fly ash, exhaust gases and waste water was measured. They recovered 39% and 33% of the cerium oxide in each test, with the proportion of recovered cerium oxide in the exhaust gases being respectively undetectable and 0.0001%. The authors note that because of their large surface to volume ratios, nanoparticles tend to adhere to surfaces within the plant resulting in an increased travel time through the system.

4.4 Effects of Control Devices upon Particle Emissions

Amongst particle control techniques Holder et al. (2013) identify older technologies of cyclones and wet scrubbers (the latter primarily used to treat gases although they will also remove some particles), and more modern techniques of dry and wet electrostatic precipitators and fabric filters.

4.4.1 Electrostatic precipitators and cyclones

The effect of switching a wet electrostatic precipitator (that was located on a MSW incinerator after a fabric filter) on and off was investigated by Maguhn et al. (2003) who found that the precipitator achieved a one order of magnitude reduction in particulate number concentration at particle

406 diameters greater than about 0.05 µm, with reduced efficiency at smaller diameters. Bologa et al.
407 (2012) found mass collection efficiencies of electrostatic precipitators on biomass combustion
408 facilities of 82% to 87%. In tests on electrostatic precipitator trial rigs, Huang and Chen (2002)
409 found that at smaller particle diameters (< 20 or 50 nm depending upon the equipment
410 configuration) efficiencies were reduced, due to some particles not being charged. Such tests are
411 not, however, directly relevant to modern municipal waste incineration plant employing fabric
412 filters.
413
414 **4.4.2 Fabric filters**
415 The penetration of particles of any size range through a filter is defined as the ratio of the
416 concentration at the outlet (X_o) to that at the inlet (X_i) of those particles (Lind et al, 2007). The
417 collection efficiency (E) of the filter at that size range can then be defined as; $E = (1$
418 $- X_o) / X_i$.
419
420 Lind et al. (2007), Wilen et al. (2007) and Zeuthen et al. (2007) measured particle concentrations
421 before and after the fabric filters of MSW incineration plants and report the percentage of particles
422 penetrating the filter over a range of particle sizes. Buonanno et al. (2010) report the particle size
423 distributions measured before and after the fabric filters of two MSW incineration plants. The
424 collection efficiency spectra (collection efficiency = 1 – penetration) were calculated from the
425 graphs in these papers, and are presented in Figure 4. There are strong similarities in the
426 descriptions of one of the plants investigated by Lind et al. (2007) and that investigated by Wilen et
427 al. (2007) and it may be the same plant, but because of the difference in collection efficiency at
428 larger particle diameters the results of both papers are shown in Figure 4. There is considerable
429 similarity between the results of the different authors with minima in the collection efficiency
430 spectra at diameters around 0.04 µm and between 0.7 and 1.5 µm.
431

Zeuthen et al. (2007) ascribe the efficiency reduction at larger diameters (0.7 – 1.5 µm) to the point of minimum efficiency of the fabric filter. They suggest that the minimum efficiency at around 0.04 µm is due to the evaporation of wet droplets in a wet scrubber which followed the fabric filter in the plant that they examined, a problem also identified by Maguhn et al. (2003) and by Ozgen et al. (2012) who measured higher particle number concentrations on a plant with a wet filtration system. Ozgen et al. (2015) measured the emissions from a MSW incinerator which had parallel wet and dry air pollution control systems, and found that the particle number concentration in the wet line was twice that in the dry line. They suggest that the higher moisture content of the flue gas in the wet line resulted in greater nucleation of gaseous precursors such as sulphuric acid and ammonia. Such scrubbers are not however reported as being present in the other plants (Lind et al., 2007; Wilen et al., 2007; Buonanno et al., 2012) which also show a minimum collection efficiency at particle diameters around 0.04 µm (Figure 4).

This minimum calculated collection efficiency is at a particle diameter where number concentrations were low and this may affect the accuracy of the calculation of collection efficiency at this diameter. Lind et al. (2007) note the difficulty in making measurements due to the low concentrations after the fabric filter. Buonanno et al. (2012) note that at particle diameters where concentrations were highest, higher collection efficiencies resulted in high overall collection efficiencies.

4.5 Comparison of Incinerator Emissions with Ambient Concentration Measurements

Hourly measurements of ambient particle number concentrations have been made by condensation particle counter for a number of years at roadside and background sites in London and at a rural site at Harwell (NPL, 2013). The annual mean particle number concentrations measured in the four years between 2008 and 2011 (following changes to the permissible concentration of sulphur in diesel fuel in late 2007 - Jones et al., 2012) and are presented in Table 2 to allow comparison with

458 the particle number concentrations measured in the flues of MSW incinerators presented in Table 1.
459 These are relevant comparators as the largest UK municipal waste incinerator (Edmonton) is
460 located in London, and others are within suburban or rural areas of the country. The values of
461 ambient particle number concentration at the background and roadside sites in London (Table 2) are
462 generally similar to, and in some cases greater than, the particle number concentrations measured in
463 the exhaust emissions from MSW incinerators listed in Table 1. At their first three sites (Table 1, F,
464 G and H), Cernuschi et al. (2012) measured ambient particle number concentrations in the vicinity
465 of the plant of 3.2×10^4, 1.4×10^4 and 2×10^4 respectively. In the case of the first two plants the
466 particle number concentrations emitted from the MSW incinerators are lower than the particle
467 number concentrations that the authors measured in the ambient air.
468
469 Particle mass and number concentrations and composition were measured at a location 200 m from
470 a MSW incinerator and 400 m from a six lane highway by Buonanno et al. (2010). They conclude
471 that the major source of locally produced particulate matter was the highway, with most of the
472 elemental composition measurements being attributed to long range atmospheric transport.
473 Buonanno et al. (2009) use published emission factors for particle numbers from road traffic to
474 compare the quantity of traffic that would produce the same particle emissions as a MSW
475 incinerator. They concluded that incinerator emissions over the course of one hour are equivalent to
476 20 vehicles (6% to 8% heavy duty) moving along 3 km of highway in typical traffic conditions. It
477 should be noted, however, that this calculation is based on emission factors from vehicles which
478 predate the reduction in the permitted quantity of sulphur in diesel fuel, which resulted in a 60%
479 reduction in particle number emissions compared to NO_x emissions from vehicles in the UK in
480 November 2007 (Jones et al., 2012).
481
482 Buonanno and Morawska (2014) found the median particle number concentration in the emissions
483 from a MSW incinerator stack to be less than the median background concentrations in the local

484 community, and substantially less that the median concentrations indoors which contribute a large
485 part of the particulate number dose which the population receives. Modelling of the dispersion of
486 particulate matter from a MSW incinerator (Scurigio et al., 2015) showed that the most significant
487 determinant of concentration at ground level was the efficiency of the flue gas treatment system.
488 Font et al. (2015) found some mass ratios of trace metals typical of those emitted by MSW
489 incinerators near to two out of six of the UK plants that they studied, while concluding that MSW
490 incinerators contributed little to ambient PM_{10} concentrations.
491
492 Ragazzi et al. (2013) report particle number concentration distributions at the exhaust stack of a
493 MSW incinerator, and at various rural and developed locations around it. At the incinerator exhaust
494 the number size distribution had a broad but dominant mode around 75 nm diameter. At the
495 surrounding locations, the particle number distributions were dominated by particles of less than 25
496 nm diameter, but with a subsidiary mode at around 75 nm at the site closest to the plant, where
497 overall concentrations were higher than in the exhaust. A time series of concentrations of particle
498 number and NO_x in ambient air at one of the sites close to the incineration plant showed a strong
499 correlation between the two determinands which the authors attributed to emissions of both
500 pollutants from road traffic. Their results failed to show any significant influence of the
501 incineration plant upon local concentrations of ultrafine particles.
502
503 The particle number size spectra of emissions from MSW, biomass, gas and gas oil powered
504 combined heat and power plants were measured by Fuglsang et al (2010) and are compared in
505 Figure 5. The emissions from the MSW incinerator with fabric filter (WTE2) are notable for having
506 relatively low concentrations at smaller particle sizes and relatively high concentrations at particle
507 diameters around 0.5 µm. In contrast, the biomass fuelled plant (with electrostatic filtration) and the
508 gas and gas oil fired plants have emissions with maximum concentrations at particle diameters less

than 0.1 µm. Fuglsang et al. (2010) suggest that the higher emissions of ultrafine particles from the gas fired plant are probably due to the use of lubrication oil in the engine in which the gas is burnt.

5. CONCLUSION

There are only a rather modest number of studies of ultrafine particle emissions from municipal waste incinerators, the majority from plants in Italy and Scandinavia. These paint a consistent picture of fabric filter abatement devices, which are used in all UK plants, working with very high efficiency, and average concentrations of particle number (representative of ultrafine particle concentrations) in the typical range of 10^3 to 10^5 cm^{-3}. Such concentrations are typical of those in urban air and hence after dispersion of the exhaust gases, concentrations of ultrafine particulate in ambient air will be dominated by those in the background air into which the incinerator gases are mixed. In some cases the flue gas concentrations of ultrafine particles fall below those in the local ambient air.

Those studies which have included measurements within ambient air in the vicinity of incinerators confirm that the incinerator emissions are not impacting significantly upon concentrations of ultrafine particles in the locality which tend to be dominated by sources such as road traffic and domestic combustion, as is the case in other localities.

While the studies of the efficiency of fabric filters on MSW incinerators show lower collection efficiencies at particle diameters around 0.04 µm, this may be a consequence of difficulties in quantifying the small number of particles of this diameter. The sole identified experimental measurement of the release of engineered nanoparticles from an MSW incinerator shows the concentration of particles released to be negligible.

The low emissions of ultrafine particles from MSW incinerators can be attributed to the introduction of material to absorb gaseous emissions in the abatement plant, which restricts the formation of ultrafine particles within the plant, and the use of high efficiency filtration at the end of the process. While no ultrafine emissions data for MSW incinerators in the UK was identified, the findings of low emissions of ultrafine particulate matter was common to plants in a number of EU countries subject to the same regulatory regime and general operating procedures as plants in the UK. While plants across the EU are operated within the same regulatory framework, there may be differences in operational practice, or waste content, between countries. The possibility that there may be an effect of any minor differences in operating procedure or waste content between the UK and other EU countries could only be determined by measurements on a UK plant.

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767

TABLE LEGNEDS

Table 1: Operating conditions, particle number measurement procedures, and particle number concentrations from the studies identified in the literature.

Table 2: Mean annual particle number concentrations measured at two London sites and Harwell rural site following the mandatory sale of "sulphur free diesel" for use in road vehicles in November 2007 (from NPL, 2013; Figure 4-36).

FIGURE LEGENDS

Figure 1: Example of ambient particle number and mass size distributions in rural air (from Harwell site, 25 July 2012).

Figure 2: Schematic diagram of a MSW incinerator.

Figure 3: Particle number concentration distributions of the emissions reported by various authors from different MSW incinerators. (Dotted lines indicate ± 1 s.d. for Zeuthen et al. 2007).

Figure 4: Particle collection efficiency spectra for fabric filters as a function of particle diameter measured at MSW incinerators by various authors. (Dotted lines indicate ± 1 s.d. for Zeuthen et al. 2007).

Figure 5: Particle number distributions for Combined Heat and Power plants using solid waste incineration (WTE 2), biogas (GF2), biomass (wood chips and saw dust) (BM 3) and gas oil (GO) (Fuglsang et al. 2010).

Table 1: Operating conditions, particle number measurement procedures, and particle number concentrations from the studies identified in the literature.

AUTHOR (Lead)	FUEL Type	FUEL Rate	FUR -NACE TYPE	EMISSION CONTROLS	SAMPLING SYSTEM	SIZE RANGE (nm)	MASS CONCENTRATION	NUMBER CONCENTRATION EMITTED (cm^{-3})		MODE IN NUMBER DISTRIBUTION (nm)	Sample temperature (°C)	Dilution factor
								Median (or mean) (cm^{-3})	1st-3rd quartiles (cm^{-3})			
Buonanno (2009)		2x7.5 Mg hr^{-1}	MG	SNCR (urea); ESP; sprayA ($NaHCO_3$, AC); FF	ELPI Dekati	30-2000	0.22 (TSP)	$1\text{-}2\times10^5$		80	0-350	1:20 to 1:200
Buonanno (1) (2011, 2012)	RDF	10-12 Mg hr^{-1}	MG	SNCR; sprayA(CaO, AC, urea); FF	SMPS 3936 CPC 3775	6-800 > 4	0.68 (TSP)	3.5×10^2	$1.5\text{-}5\times10^2$	60-100 (at max emis)	120-150	1:25
Buonanno (2) (2012)	RDF	15 Mg hr^{-1}	MG	SNR, sprayA($NaHCO_3$, AC); FF	SMPS 3936 CPC 3775	6-800 > 4	1 (TSP)	1.5×10^2	$0.1\text{-}1\times10^3$	15, 200 (at max emis)	120-150	1:25
Buonanno (3) (2012)	MSW	9-10 Mg hr^{-1}	RG	wet SCR($Ca(OH)_2$, NaOH soln, NH_3 soln); FF	SMPS 3936, Grimm, CPC 3775, CPC 5403	5.5-800 > 4	2 (TSP)	3×10^3	$1\text{-}7\times10^3$	60-100 (at max emis)	150	1:7
Buonanno (4) (2012)	MSW	12.5 Mg hr^{-1}	MG	$Ca(OH)_2$ injection; FF; ($NaHCO_3$, AC, NH_3 soln); FF	Grimm DMA 55706, CPC 5403 (Grimm)	5.5-350 > 4.5	0.9 (TSP)	6×10^2	$0.3\text{-}1.2\times10^3$	8, 30 (at max emis)	150	1:4.5
Cernuschi (1) (2012)	urban waste	900-1200 Mg d^{-1}	MG	ESP, dryA($NaHCO_3$+AC), FF(180-190°C), SCR(180°C)	ELPI Dekati	7-2500	n.a.	1.6×10^4 [$\pm3.9\times10^3$]	$0.45\text{-}3.0\times10^4$	32	24 to 31	1:40 to 1:50
Cernuschi (2) (2012)	urban waste	650-1200 Mg d^{-1}	MG	SCR(250°C), drA(lime+AC), FF(130-140°C)	ELPI Dekati	7-2500	n.a.	7.9×10^3 [$\pm2.5\times10^3$]	$0.2\text{-}1.6\times10^4$	25	24 to 31	1:40 to 1:50
Cernuschi (3) (2012)	urban waste	600-700 Mg d^{-1}	MG	Quencher, dryA(Sorbalit+AC), FF(150^{0}C), WA(water+NaOH sol), SCR(250°C)	ELPI Dekati	7-2500	n.a.	7.8×10^4 [$\pm1.4\times10^4$]	$0.48\text{-}1.3\times10^5$	17, 81	24 to 31	1:40 to 1:50
Cernuschi (4) (2012)	urban waste	200 Mg d^{-1}	MG	SNCR(water+urea post comb), dryA(sodium bicarbonate+AC), FF, wetA	ELPI Dekati	7-2500	n.a.	5.7×10^5 [$\pm2.9\times10^5$]	$5.5\text{-}31\times10^5$	19	24 to 31	1:20 to 1:40
Maguhn (2003)	MSW	23 MW		sprayAA; FF, acid/basic wet scrubber; wet ESP	SMPS 3071 /3022 and APS 3310,	17-30000	n.a.	1.8×10^5 (from Fig 8a)		70 (<40 inter-mittent)	80	1:10 to 1:10000
Zeuthen (2007)	MSW	22 MW	grate	lime, FF, WS	SMPS 3071/3010	14-800	n.a.	6.9×10^4		167	-	1:5 to 1:200
Wilen (2007)	waste	75 MW	CFBR	AC, lime, FF	ELPI, TEOM	-	0.2-0.7 (TSP)	approx 2×10^4		-	20	"heavily diluted"
FORCE (WTE2) (2009)	waste	<30 MWe	grate	SNCR, semi-dry CaO, AC, FF	ELPI	7-2500	2.2 (PM2.5)	1.7×10^4		500	25	1:10
FORCE (WTE3) (2009)	waste	<30MWe	grate	SNCR, ESP, Scrubber + CaO, CaO + AC, FF	ELPI	7-2500	0.02 (PM2.5)	5.9×10^1		30-50	25	1:10
Ozgen (1)	waste			dry technology for	ELPI	7-2500	n.a.	$1.1\text{-}1.7\times10^4$		n.a.	n.a.	n.a.

(2012)				particulate								
Ozgen (2) (2012)	waste			dry technology for particulate	ELPI	7-2500	n.a.	$5\text{-}8\times10^{3}$		n.a.	n.a.	n.a.
Ozgen (3) (2012)	waste			Dry/wet technology for particulate	ELPI	7-2500	n.a.	$4\text{-}8\times10^{4}$		n.a.	n.a.	n.a.

Abbreviations:

A – absorption
AC – activated carbon
APS – aerodynamic particle sizer
CPC – condensation particle counter
DMA – differential mobility analyser
ELPI – electrical low pressure impactor
ESP – electrostatic precipitator
FBR – fluidised bed
CFBR – circulating fluidised bed
FF – fabric filter
MG – moving grate
MSW – municipal solid waste
$PM_{2.5}$ – particulate matter less than 2.5 µm aerodynamic diameter
RDF – refuse derived fuel
RG – rotating grate
SMPS - scanning mobility particle sizer
S(N)CR – selective (non-)catalytic reduction
TEOM – tapered element oscillating microbalance
TSP - total suspended particulate

Table 2: Mean annual particle number concentrations measured at two London sites and Harwell rural site following the mandatory sale of "sulphur free diesel" for use in road vehicles in November 2007 (from NPL, 2013; Figure 4-36).

Year	Marylebone Road (roadside site)	North Kensington (urban background site)	Harwell (rural site)
2008	3.6×10^4	1.35×10^4	4.7×10^3
2009	3.5×10^4	1.40×10^4	5.6×10^3
2010	3.8×10^4	1.40×10^4	5.5×10^3
2011	3.4×10^4	1.35×10^4	4.3×10^3

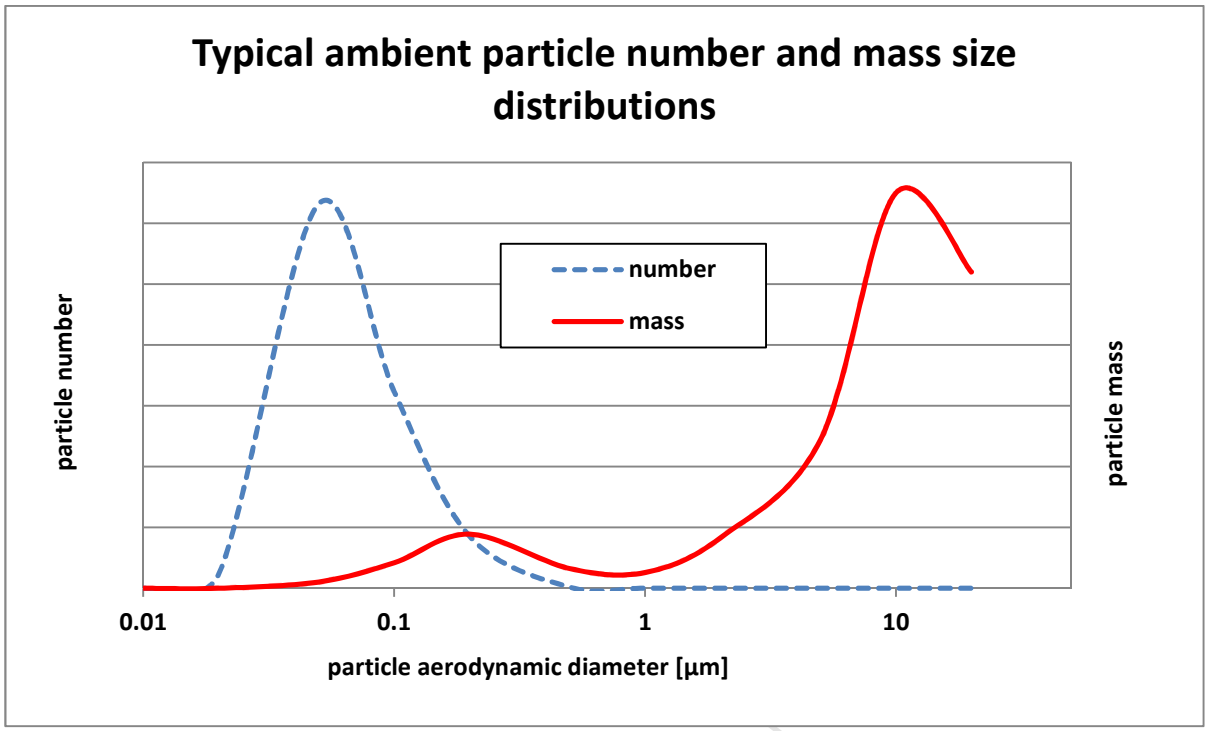

Figure 1: Example of ambient particle number and mass size distributions in rural air (data from Harwell site, 25 July 2012, [https://uk-air.defra.gov.uk/data]).

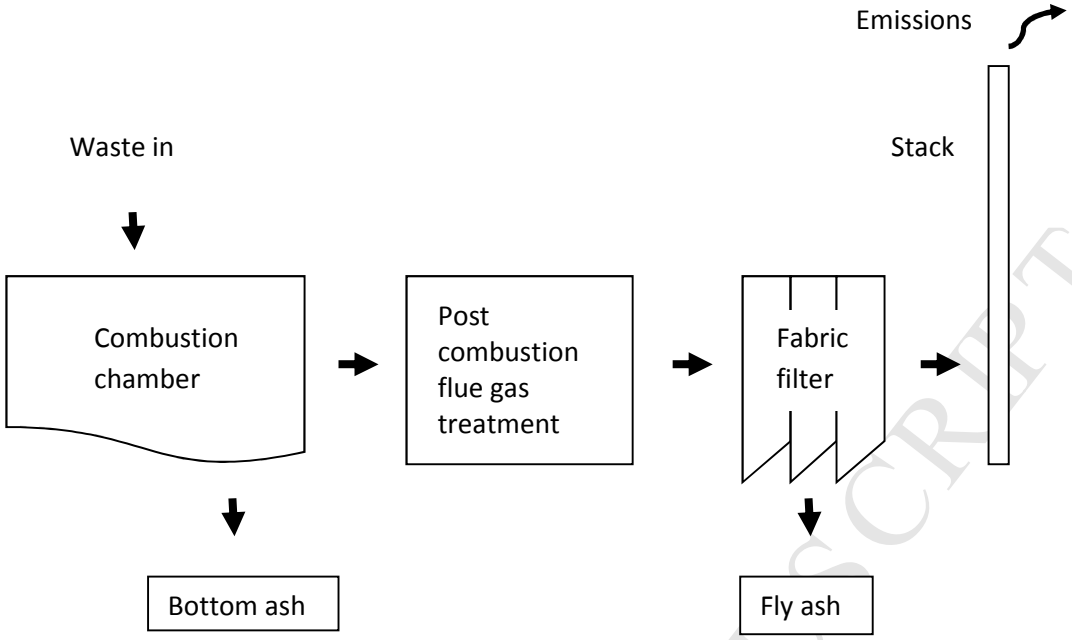

Figure 2: Schematic diagram of a MSW incinerator.

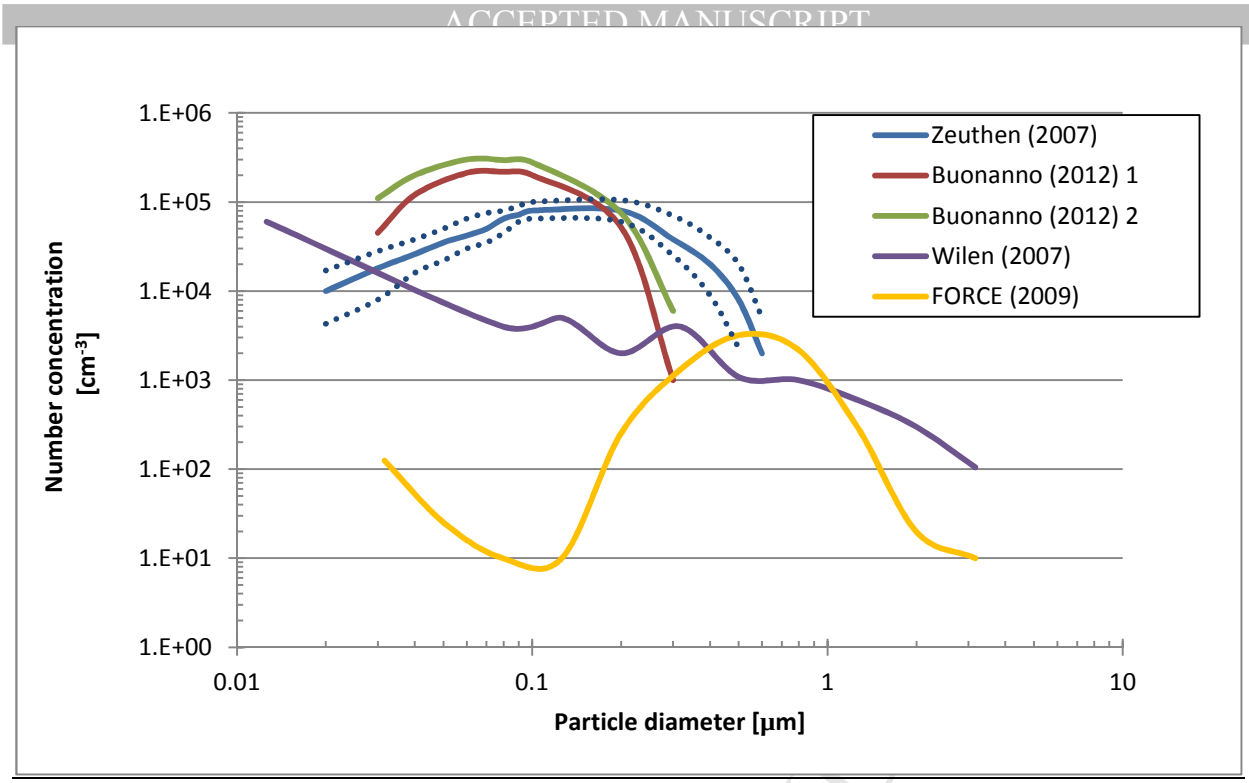

Figure 3: Particle number concentration distributions of the emissions reported by various authors from different MSW incinerators. (Dotted lines indicate ± 1 s.d. for Zeuthen et al. 2007).

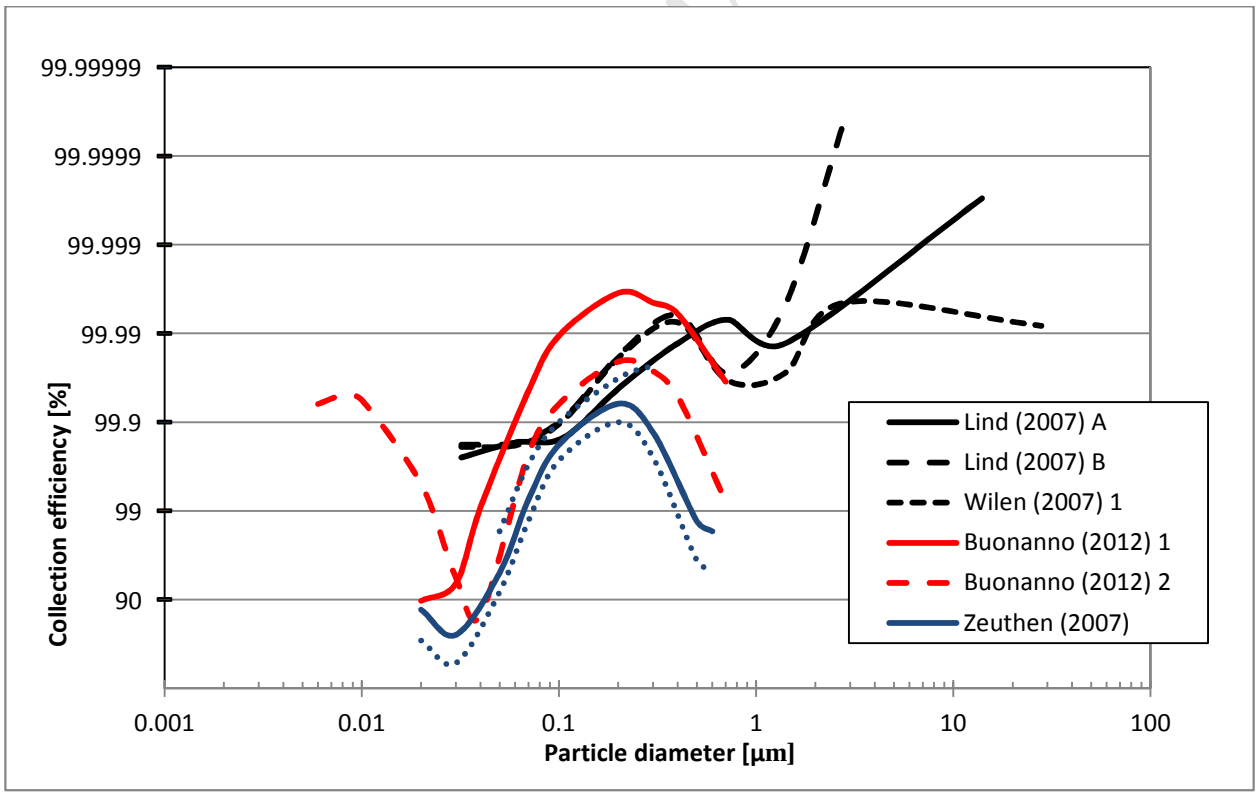

Figure 4: Particle collection efficiency spectra for fabric filters as a function of particle diameter measured at MSW incinerators by various authors. (Dotted lines indicate ± 1 s.d. for Zeuthen et al. 2007).

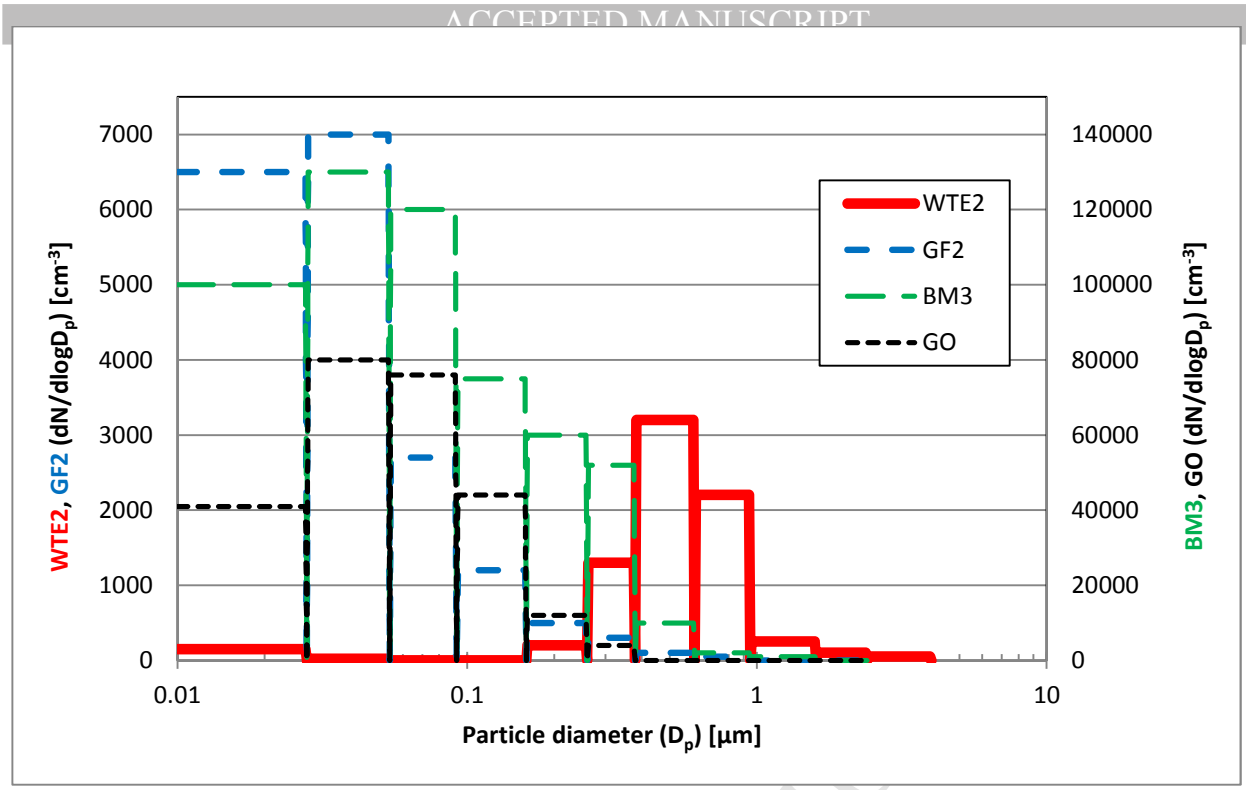

Figure 5: Particle number distributions for Combined Heat and Power plants using solid waste incineration (WTE 2), biogas (GF2), biomass (wood chips and saw dust) (BM 3) and gas oil (GO) (Fuglsang et al. 2010).